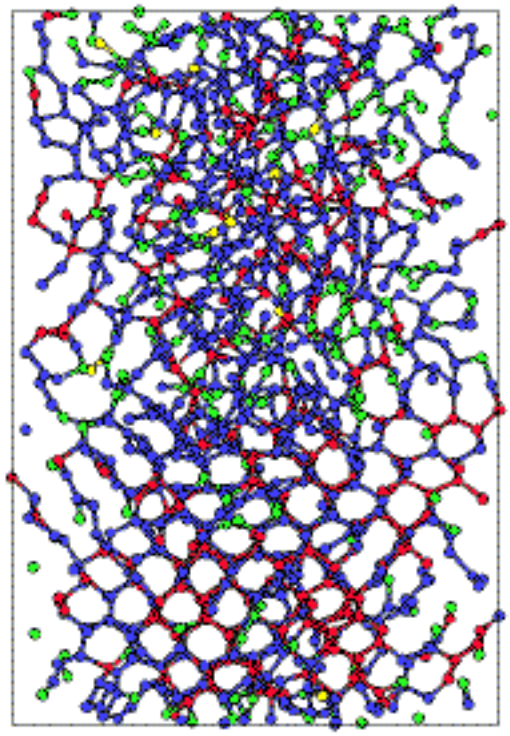
VI.B Melting A Diamond Crystal with Tight Binding Molecular Dynamics

Introduction: Well beyond its value for jewelry, diamond is invaluable as the hardest known substance. Fine diamond particles are the ultimate abrasive, and wear resistant diamond coatings are used on tools from saw blades to surgical instruments. But who would consider melting diamond? Materials scientists and engineers who are searching for cheaper and more robust processing routes to the synthesis of diamond coatings could use the thermodynamic phase diagram of carbon as a guide. The conditions that it takes to melt diamond are too extreme for careful laboratory experimentation. Melting diamond on a computer, however, makes it possible to determine the complete pressure-temperature phase diagram.



Calculational Notes: Quantum mechanical tight binding (TB) molecular dynamics calculations were run with a parallel code and an algorithm that scales linearly with the system size, for large systems. A highly efficient implementation permits the calculation of the electronic structure and forces for systems of up to 10,000 atoms on the 1024 node Paragon XP/S 150 at ORNL. The empirical parameters for the TB code were determined by fits to the results from extensive first principles calculations for many static geometries.

Results: The figure shows a snapshot of 2000 carbon atoms (T>6000 K) with the lower half in a diamond lattice and the upper half in a liquid phase. Red atoms indicate four-fold bonded (diamond-like) atoms; the blue atoms indicate three-fold (graphitic) bonded atoms; and there exist some two-fold and five-fold coordinated atoms. The large number of three-fold atoms is an indication that the liquid phase is less dense than the four-fold diamond phase. By changing temperature and/or pressure, the interface between the solid and liquid will move (e.g., the solid fraction increases below the melting point). Such simulations permit the accurate determination of the melting temperature of diamond as a function of pressure.

Significance: Molecular dynamics simulations can lead to new insights for understanding natural and artificial diamond synthesis. Experiments that are difficult, expensive, or impossible in the laboratory, can be accurately simulated on modern computers. By including the essential quantum nature of the electrons and their chemical bonds, the tight binding calculations form a natural bridge leading from first principles electronic structure calculations to the mesoscopic regime. This eliminates the need to develop new force fields or other more simplified interactions among atoms in complex environments. SSI resources will permit the extension of the quantum mechanical treatment to even larger numbers of atoms in order to investigate more complex nanoscale phenomena involving extended defects. This would illuminate the fundamental relationship between microstructure and important macroscopic materials properties.